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13. ABSTRACT *(Maximum 200 Words)*

First-principles calculations were performed of a variety of materials systems of potential interest in spintronics. A comprehensive study was made of transitional metal doping in SiC and their magnetic properties. The trends in magnetic properties and in the preference for rocksalt versus zincblende structure were studied for the entire series of transition metal nitrides. The LSDA+U method was implemented in the FP-LMTO approach and applied to rare-earth nitrides and related compounds. A study was made of half-metallicity in zincblende transition metal compounds. A study was made of Mn doping of ScN. It was found to be a potentially interesting dilute magnetic semiconductor system. Exchange interactions in this system were calculated using a linear response approacha and cluster variation method calculations indicate a Tc above room temperature should be feasible in this material. Calculations of the optical spectra of antifeorromagnetic MnN were compared with experimental data.

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Magneto-Optical Properties of Hybrid Magnetic Material Semiconductor Nanostructures

Walter R. L. Lambrecht

September 14, 2007

1 Publications

a. Publications published in peer reviewed journals

1. *Magnetic properties of substitutional 3d transition metal impurities in silicon carbide*, M. S. Miao and Walter R. Lambrecht, Phys. Rev. B **68**, 125204 (2003)

2. *Electronic structure and magnetic interactions in MnN and Mn_3N_2*, Walter R. Lambrecht, Margarita Prikhodko, and M. S. Miao, Phys. Rev. B **68**, 174411 (2003)

3. *First-principles study of the preference for zinc-blende or rocksalt structures in FeN and CoN*, Pavel Lukashev and Walter R. L. Lambrecht, Phys. Rev. B **70**, 245205 (2004)

4. *Stability and half-metallicity of transition metal pnictides in tetrahedrally bonded structures*, M. S. Miao and Walter R. L. Lambrecht, Phys. Rev. B **71**, 064407 (2005)

5. *Structure and magnetic properties of MnN, CrN, and VN under volume expansion*, M. S. Miao and Walter R. Lambrecht, Phys. Rev. B **71**, 214405 (2005)

6. *Electronic structure and magnetic properties of Mn_3GaN precipitates in $Ga_{1-x}Mn_xN$*, M. S. Miao, Aditi Herwadkar, and Walter R. Lambrecht, Phys. Rev. B **72**, 033204 (2005)

7. *Effects of biaxial strain on stability and half-metallicity of Cr and Mn pnictides and chalcogenides in the zinc-blende structure*, M. S. Miao and Walter R. Lambrecht, Phys. Rev. B **72**, 064409 (2005)

8. *Optical conductivity and x-ray absorption and emission study of the band structure of MnN films*, S. Granville, B. J. Ruck, F. Budde, A. Koo, J. E. Downes, H. J. Trodahl, A. Bittar, N. Strickland, G. V. Williams, W. R. Lambrecht, Timothy Learmonth, Kevin E. Smith, V. J. Kennedy, A. Markwitz, and Thorsten Schmitt, Phys. Rev. B **72**, 205127 (2005)

9. *Mn-doped ScN : A dilute ferromagnetic semiconductor with local exchange coupling*, Aditi Herwadkar and Walter R. L. Lambrecht, Phys. Rev. B **72**, 235207 (2005)

10. *Electronic driving force for stacking fault expansion in 4H–SiC*, Walter R. L. Lambrecht and M. S. Miao, Phys. Rev. B **73**, 155312 (2006)

11. *Electronic structure of Gd pnictides calculated within the LSDA+U approach*, P. Larson and Walter R. L. Lambrecht, Phys. Rev. B **74**, 085108 (2006)

12. *Electronic structure and magnetic properties of transition-metal-doped 3C and 4H silicon carbide*, M. S. Miao and Walter R. Lambrecht, Phys. Rev. B **74**, 235218 (2006)

13. *Electronic structure of rare-earth nitrides using the LSDA+U approach: Importance of allowing 4f orbitals to break the cubic crystal symmetry*, P. Larson, Walter R. L. Lambrecht, Athanasios Chantis, and Mark van Schilfgaarde, Phys. Rev. B **75**, 045114 (2007)

14. *The effects of biaxial strain on stability and half-metallicity of zinc blende CrSb*, M. S. Miao and Walter R. L. Lambrecht, J. Appl. Phys. **97**, 10C304 (2005)

15. *First-principles study of the structural and magnetic properties of iron indium nitride*, Pavel Lukashev and Walter R. L. Lambrecht J. Appl. Phys. **97**, 10D309 (2005)

16. Magnetic properties of transition-metal nitrides Walter R. L. Lambrecht, M. S. Miao, and Pavel Lukashev J. Appl. Phys. **97**, 10D306 (2005)

17. *Atomic-Scale Spin-Polarized Scanning Tunneling Microscopy Applied to $Mn_3N_2(010)$*, Haiqiang Yang, Arthur Smith, Margarita Prikhodko, and Walter R. L. Lambrecht, Phys. Rev. Lett. **89**, 226101 (2002)

18. *Atomic-Scale Structure of η-phase Mn_3N_2 (010) Studied by in-Situ Scanning Tunneling Microscopy and First-Principles Theory*, Haiqiang Yang, Rong Yang, Arthur R. Smith, and Walter R. L. Lambrecht, Surf. Sci. **548**, 117-128 (2003)

19. *Aspects of Spin-Polarized Scanning Tunneling Microscopy at the Atomic Scale: Experiment, Theory, and Simulation*, Arthur R. Smith, Rong Yang, Haiqiang Yang, and Walter R. L. Lambrecht, Surface Science **561** (2-3), 154 (2004)

20. *Crystal Structure, Electronic Structure and Magnetism of Transition Metal Nitrides*, M. S. Miao, Pavel Lukashev, Aditi Herwadkar, and Walter R.L. Lambrecht, Physica Status Solidi (c) **2**, 2516-2519 (2005)

21. *Recent advances in atomic-scale spin-polarized scanning tunneling microscopy*, A. R. Smith, R. Yang, H. Q. Yang, A. Dick, J. Neugebauer, and W. R. L. Lambrecht, Microscopy Research and Technique **66**, 72-84 (2005)

22. *Electronic structure and magnetism of europium-chalcogenides in comparison with gadolinium nitride*, P. Larson and W. R. L. Lambrecht, J. Phys. Condens. Matter **18**, 11333-11345 (2006)

b. Manuscripts submitted to peer reviewed journals but not yet published

1. *Linear-response study of the exchange interactions in Mn-doped ScN: effects of disorder, band gap and doping*, Aditi Herwadkar, Walter R. L. Lambrecht, and M. van Schilfdgaarde, submitted to Phys. Rev. Lett. (2007)

c. Publications in conference proceedings

1. *Electronic Structure and Magnetic Properties of Transition Metal Doped Silicon Carbide*, M.S. Miao and Walter R.L. Lambrecht, in *Spintronics*, edited by T. J. Klemmer, J. Z. Sun, A. Fert, and J. Bass, Mater Res. Soc. Symp. Proc. 690, F6.8 (2002)

d. Presentations at conferences not published in proceedings

1. *Atomic-Scale Spin-Polarized Scanning Tunneling Microscopy Studies of the Surface Magnetic Structure of $Mn_3N_2(010)$*, Haiqiang Yang, Arthur R. Smith, Margarita Prikhodko, and Walter R. L. Lambrecht, Bull. Am. Phys. Soc. **48**, N14.009 (2003),

2. *Bias-Voltage Dependent Magnetic Contrast in Spin-Polarized Scanning Tunneling Microscopy*, Rong Yang, Haiqiang Yang, Arthur R. Smith, Margarita Prikhodko, and Walter R. L. Lambrecht, Bull. Am. Phys. Soc.**48**, N14.010 (2003).

3. *New Materials for Spintronics and Nonlinear Optics*, M. S. Miao, X. Jiang and W. R. L. Lambrecht, Research Showcase 2003, CWRU, Cleveland, OH April 4, 2003

4. *First-principles study of CoN and FeN*, Pavel Lukashev and W. R. L. Lambrecht Fall Meeting of the Ohio Section October 17-18, 2003 Case Western Reserve University Cleveland, Ohio, Abstract P.001

5. *Zincblende versus Rocksalt Structure in CoN and FeN*, Pavel Lukashev and Walter R. L. Lambrecht APS March meeting 2004 (Montreal), abstract B24.006 Bull. Am. Phys. Soc. **49** (2004)

6. *Electronic structure and magnetism in Mn_4N and Mn_2N*, Aditi Herwadkar and Walter R. L. Lambrecht, APS March meeting 2004 (Montreal), abstract D24.009 Bull. Am. Phys. Soc. **49** (2004)

7. *Magnetic Interactions in MnN and Mn_3N_2*, Walter R. L. Lambrecht, Margarita Prikhodko and Maosheng Miao APS March meeting 2004 (Montreal), abstract D24.010 Bull. Am. Phys. Soc. **49** (2004)

8. *Electronic Structure of GdN: Magnetic and Structural Properties*, Paul Larson and Walter R. L. Lambrecht, APS March meeting 2004 (Montreal), abstract D24.011 Bull. Am. Phys. Soc. **49** (2004)

9. *Crystal Structure, Electronic Structure and Magnetism of Transition Metal Nitrides*, Maosheng Miao, Paul Larson, Pavel Lukashev, Aditi Herwadkar and W. R. L. Lambrecht, International Workshop on Nitride Semiconductors, July 19-23, 2004 Pittsburg, PA

10. *Bias-Voltage-Dependent Magnetic and Non-magnetic Corrugation in Atomic Scale Spin Polarized Scanning Tunneling Microsopy*, Arthur R. Smith, Rong Yang, Haiqiang Yang and Walter R. L. Lambrecht, Mater. Res. Soc. Meeting Boston December 2-3 2003, Symposium GG: Advanced Characterization techniques for Data Storage Materials

11. *Magnetic properties of transition metal nitrides,* Walter R. L. Lambrecht, M.S. Miao, and Pavel Lukashev, 49th Annual Conference on Magnetism and Magnetic Materials, Jacksonville, Florida November 7-11, 2004.

12. *First-principles Study of the Structural and Magnetic Properties of Iron Indium Nitride,* Pavel Lukashev and Walter R. L. Lambrecht, 49th Annual Conference on Magnetism and Magnetic Materials, Jacksonville,Florida November 7-11, 2004

13. *The effects of biaxial strain on stability and half-metallicity of transition metal pnictides and chalopyrites* in zinc blende structure, M. Miao and Walter R. L. Lambrecht, 49th Annual Conference on Magnetism and Magnetic Materials, Jacksonville, Florida November 7-11, 2004.

14. *Electronic structure of Mn-doped ScN: a possible magnetic semiconductor,* A. A. Herwadkar and W. Lambrecht, 49th Annual Conference on Magnetism and Magnetic Materials, Jacksonville, Florida November 7-11, 2004.

15. *Electronic structure and magnetism in some transition metal and rare-earth nitrides,* Pavel Lukashev, Aditi Herwadkar, Paul Larson, M. S. Miao and W. R.L. Lambrecht, Research Showcase 2004, April 2, 2004

16. *Rocksalt MnN: A Vacancy Stabilized Structure,* Maosheng Miao and Walter Lambrecht, APS March meeting, Los Angeles, March 2005, Bull. Am. Phys. Soc. **50** Abstract D9.00012

17. *Electronic structure and magnetic properties of transition-metal doped Bi_2Te_3, Bi_2Se_3, and Sb_2Te_3 for diluted magnetic semiconductors,* Paul Larson and Walter R. L. lambrecht, APS March Meeting, Los Angeles, March 2005, Bull. Am. Phys. Soc. **50** Abstract L10.00012

18. *First-principles Study of the Structural and Magnetic Properties of Cobalt Indium Nitride,* Pavel Lukashev and Walter R. L. Lambrecht, APS March Meeting, Los Angeles, March 2005, Bull. Am Phys. Soc. **50** Abstract H10.00009

19. *ScN:Mn a dilute ferromagnetic semiconductor,* Aditi Herwadkar and Walter R. L. Lambrecht, Bull. Am Phys. Soc. **50** Abstract H10.00008

20. *Magnetism in transition metal and rare earth nitrides using electronic structure calculations,* Aditi Herwadkar, Pavel Lukashev, Paul Lar-

son, M. S. Miao, W. R. L. Lambrecht, Showcase 2005, Case Western Reserve University, Cleveland, April 6-7 , 2005.

21. *Optical conductivity of MnN: a combined experimental and theoretical study,* W. R. L. Lambrecht, S. Granville,B. J. Ruck, F. Budde, A. Koo, J. E. Downes, H. J. Trodahl, A. Bittar, N. Strickland, G. V. M. Williams, Timothy Learmont, Kevin E. Smith, V. J. Kennedy, A. Markwitz, APS March Meeting, Baltimore 2006, abstract A15.00010, http://www.aps.org/meetings/baps/archives-2006.cfm

22. *Rare-earth nitrides: an LSDA+U study,* P. Larson, W. R. L. Lambrecht, M. van Schilfgaarde, APS March Meeting, Baltimore 2006, abstract Z23.00004, http://www.aps.org/meetings/baps/archives-2006.cfm

23. *Electronic structure of CrN: a Mott insulator,* Aditi Herwadkar, W. R. L. Lambrecht, M. van Schilfgaarde, APS March Meeting, Baltimore 2006, abstract Z23.00005, http://www.aps.org/meetings/baps/archives-2006.cfm

24. *Electronic Structure Calculations of Transition Metal and Rare Earth Nitrides, using LSDA+U,* Paul Larson, Aditi Herwadkar and W. R. L. Lambrecht, Research ShowCase , April 5-6, 2006

25. *Electronic Structure Calculations of Transition Metal and Rare Earth Nitrides, using LSDA+U,* A. Herwadkar, P. Larson and W. R. L. Lambrecht, 18th Annual Workshop on Developments in Electronic-Structure Methods, 22-25 June 2006

26. *Magnetic interactions in metallic anti-ferromagnetic nitride compounds,* Condensed Matter Seminar, Dept. of Physics, CWRU, September 8, 2003.

27. *Structural and Magnetic Properties of Transition Metal Nitrides,* University of Buffalo, December 9, 2004

28. *Transition Metal Nitrides: New Materials for Spintronics,* Walter Lambrecht, Mahidol University, Bangkok, Thailand, Jan. 14 2005

29. *Transition Metal Nitrides: New Materials for Spintronics,* Walter Lambrecht, Suranaree University of Technology, Nakhon Ratachsima, Thailand, Jan. 18 2005

30. *Transition Metal Nitrides: New Materials for Spintronics,* Walter Lambrecht, Chiang Mai University, Thailand, Jan. 25 2005

31. *Transition metal and rare-earth nitrides: a new route to magnetic semiconductors*, Walter R. L. Lambrecht, Case Western Reserve University condensed matter seminar, January 2007

32. *Transition metal and rare-earth nitrides: a new route to magnetic semiconductors*, Walter R. L. Lambrecht, Colloquium University of Toledo, February 2007

e. Manuscripts in preparation to be submitted to peer reviewed journals

1. *Electronic structure of CrN: a borderline Mott insulator*, A. Herwadkar and W. R. L. Lambrecht

2. *Rocksalt Mn, a vacancy stabilized structure*, M. S. Miao and W. R. L. Lambrecht

3. *Electronic structure of magnetism in transition-metal (Ti – Zn) doped Bi_2Te_3, Bi_2Se_3, and Sb_2Te_3*, P. Larson and W. R. L. Lambrecht

f. Theses

1. *Electronic structure and magnetism in some transition metal nitrides [electronic resource] : Mn-doped ScN, dilute magnetic semiconductor and CrN, Mott insulator*, by Aditi A. Herwadkar, Ph. D. Thesis, Case Western Reserve University, (2006), available electronically at http://www.ohiolink.edu/etd/view.cgi?acc

Summary

a. Number of published papers in peer reviewed journals: 22
b. Number of submitted (not yet published) papers to peer reviewed journals: 1
c. Number of conference proceedings published: 1
d. Number of presentations at conferences, seminars, not published: 32
e. Number of manuscripts in preparation (not yet submitted): 3
f. Number of theses completed: 1

2 Summary data on human resources

The following scientists were involved with the project:

- Walter R. L. Lambrecht, Principal Investigator, Professor of Physics

- Maosheng Miao, Postdoctoral Sr. Research Associate (part time, 2002-2006)
- Paul Larson, Postdoctoral Research Associate (full time, 2002-2005)
- Pavel Lukashev, Graduate Student (part time, completed Ph.D, 2003-2005)
- Aditi Herwadkar, Graduate Student (part time, completed Ph.D, 2003-2006)
- Margarita Prikhodko, Graduate Student (part time, 2002)

a. Number of Scientists supported by this agreement: 6
b. Number of Graduate Students supported by this agreement: 3
c. Number of Post Doctoral Associates supported by this agreement: 2
d. Number of Faculty supported by this agreement: 1
e. Number of Other Staff supported by this agreement: 0
f. Number of Undergrads supported by this agreement: 0
g. Number of PhD(s) awarded as a result of this agreement: 1
h. Number of Bachelors Degrees awarded as a result of this agreement: 0
i. Number of Master Degrees awarded as a result of this agreement: 0
j. Number of FTE graduate students supported per year: 1
k. Number of FTE postdoctoral associates supported per year: 1.5
l. Faculty summer salary support in months per year: 1

3 Report of Inventions

a. Number of inventions or discoveries disclosed to University Office of Sponsored Research: 1
Title: Mn-doped ScN, an n-type doped dilute magnetic semiconductor with Curie temperature above room temperature
Date of disclosure: September 17, 2007.
b. Number of Patents Sumbitted as a result of this agreement: 0
c. Number of Patents Awarded as a result of this agreement: 0

4 Narrative Description of Scientific Accomplishments

The main goals of this project were to study electronic structure, magnetism, and optical properties in materials with potental use in spintronics. The focus of the work was on transition metal and rare-earth nitrides. Transition metal doping in SiC was also studied.

The methodology used was based on first-principels calculations using the density functional theory (DFT) in the local spin density approximation (LSDA). During the course of the project, we developed the LSDA+U approach and implemented it in the Full-Potential Linearized Muffin-Tin Orbital method code (FP-LMTO). We also applied the linear response approach for calculation of magnetic exchange interactions based on a Green's function implementation of the atomic sphere approximation (ASA) version of the LMTO method and used no-collinear magnetic calculations in the rigid spin approximation. Optical properties of magnetic systems were calculated using the ASA version of LMTO

The main accomplishments of the project can be summarized as follows. The numbers in brackets refer to the above lists of published paper.

- A study was made of transition metal dopants in SiC. This led to two papers,[1,12] the first was restricted to 3C-SiC hosts and established the basic magnetic moment trends of the TM impurities on Si and C sites, studied the preference for ferromagnetic or antiferromagnetic coupling and discussed the competition with carbide and silicide formation. The second focused on 4H SiC and delved deeper into the energy levels of the defect, included relaxation via the FP-LMTO approach, and established the different nature of magnetic interactions in the Cr and Mn cases.

- The nature of the exchange interactions in MnN and Mn_3N_2 were studied using a mapping to a Heisenberg Hamiltonian.[2] We found it to be dominated by second nearest neighbor ferromagnetic interactions and smaller nearest neighbor antiferromagnetic interactions. The role of vacancies was studied. The calculations of the partial densities of states in Mn_3N_2 were used to assist in the interpretation of spin-polarized scanning tunneling microcopy studies by A. R. Smith at Ohio University.[17-19,21] MnN was found to actually prefer zincblende structure [e.2] but the rocksalt structure may be stabilized by a small number of N vacancies. A study of the optical interband transitions in MnN was also carried out.[8]

- A comprehensive study [3,5,15,16,20] was made of the trends of the 3d transition metal nitrides: we established their relative preference for zincblende versus rocksalt structure and the development of magnetic moments and their preferred alignment. The preference for zincblende in crystals under tensile strain was established and proposed to be relevant to the possible occurence of nanocrystalline precipitate phases in doping of GaN with transition metal nitrides. Relatedly, we also studied Mn_3GaN.[6]

- The study of transition metal nitrides was expanded to other transition metal group-V compounds.[4] A study was made of their structural preference (rocksalt, NiAs-structure, zincblende, wurtzite) as function of lattice constant, and the potentiality of achieving half-metallicity in the zincblende phase under tensile strain. Biaxial strain effects on this problem were also investigated.[7,14]

- A study was made of the possibility of using ScN as a dilute magnetic semiconductor host with doping by Mn. The rationale for this work was that alloys up to 30 % can be made because of the common crystal structure. A first study [9] using FP-LMTO established fairly strong exhange interactions and the presence of magnetic moments. A second more in-depth study was made using linear response and a more sophisticated approach to calculating transition temperatures.[b.1] Our calculations predict this system to be a ferromagnetic DMS with Curie temperature above room temperature for concentrations of about 10 % Mn. We find the system to be sensitive to doping and this may be the reason why magnetism has not yet been observed experimentally. Both n and p-type dopign reduce T_c and a background n-doping needs to stay below 10^{20}e/cm^3 to achieve magnetism.

- Our newly developed LSDA+U capability was used to study rare-earth nitrides and related compounds.[11,13,22] The symmetry breaking by development of orbital magnetic moment was found to be important in these systems. The magnetic behavior of Gd-pnictide compounds (N,P,As,Sb) and Eu-chalcogenides (EuO, EuS, EuSe, EuTe) were found to be well described by a nearest plus second nearest neighbor $S = 7/2$ Heisenberg model and our calculations allowed us to determine the exchange interactions of this model from first principles. This explains their trend in Curie-Weiss temperatures and critical magnetic fields.

- The LSDA+U approach was also used to study CrN, which was found to be at the brink of a Mott insulator transition.[e.1]
- We also started a study of a novel class of magnetic semiconductors, Bi_2Te_3, Bi_2Se_3, and Sb_2Te_3 doped with various TM atoms. These materials have been mainly studied as thermo-electrics but recently found useful as DMS hosts.[e.3]
- Unrelated to the magnetic systems but related to an earlier ONR sponsored project, we studied stacking faults in SiC.[10]

5 Interactions

Early on in the project, a collaboration was established with A. R. Smith at Ohio University. We collaborated on SPSTM of Mn_3N_2 and also exchanged ideas on Mn-doped ScN. We were contacted by a group in New Zealand, headed by J. Trodahl, to study the optical properties of MnN and subsequently various rare-earth nitrides. We continued a fruitful collaboration with Mark van Schilfgaarde, at Arizona State University. The LSDA+U was implemented in his lmf (FP-LMTO) code during a sabbatical stay of the PI at ASU. During this stay, we also became familiar with the non-collinear and linear response magnetic calculations.

6 Summary of timeline and financial report

This project funded originally at the level of $409,632 was scheduled to end on July 2006 but was extended with an additional funding of $15320 and an extension of the end date till 30-September-2006. Completing a number of the papers resulting from this work took extra time and caused the delay in this final report.